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ANALYTICAL REFERENCE SERVICE TRAINING PROGRAM. SAMPLE TYPE V-B, WATER RADIOLOGICAL

Robert A. Taft Sanitary Engineering Center
Cincinnati, Ohio

May 1961

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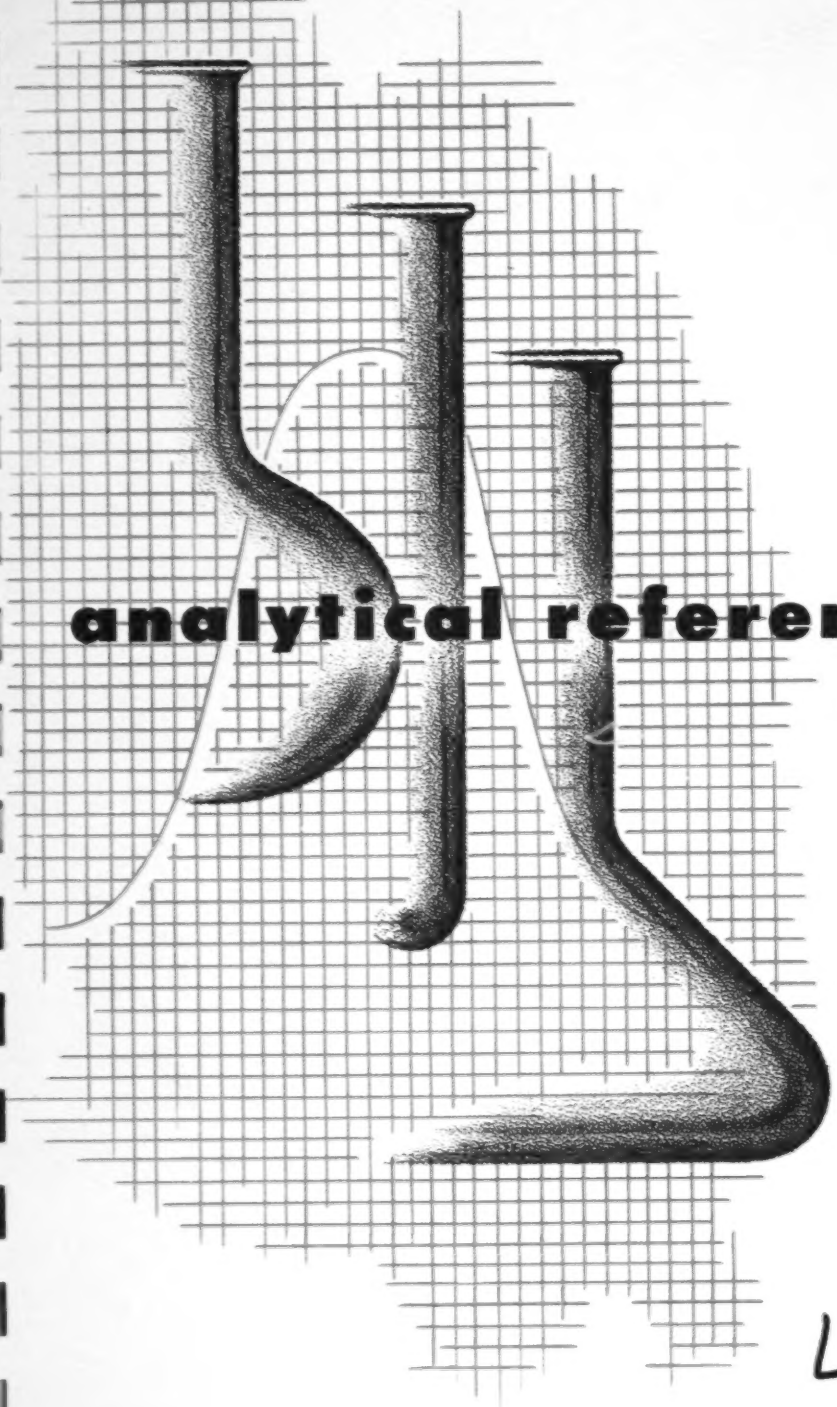

SAMPLE TYPE V-B
WATER, RADIOLOGICAL
MAY, 1961

analytical reference service

TRAINING PROGRAM

Robert A. Taft

SANITARY ENGINEERING CENTER

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
PUBLIC HEALTH SERVICE
CINCINNATI, OHIO

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UNITED STATES GOVERNMENT

PHS - DHEW

Memorandum

TO : Mr. Richard S. Green
Basic Data Branch Chief, Div. of WS & PC
Room 2501 T-R
Washington 25, D. C.

DATE: June 19, 1961

FROM : Chief, Training Program

SUBJECT: Analytical Reference Service Sample Type V-B (Water, Radiological)

A copy of the latest report of the Analytical Reference Service, Sample Type V-B, Water, Radiological, is attached. Should you desire additional copies, we will be happy to forward them.

This study, as compared to the earlier Sample Type V, Water, Radiological (1959), confirms our experience that a repeat of a study yields quite a significant improvement in accuracy and precision. Our future plans call for distribution of additional radiological samples. A sample for the identification of specific radionuclides in water will probably head the list.

It is only through the cooperative efforts of the ARS membership and guest participants that data of this type can be procured, evaluated, and serve to add to improved analytical methodology in our field.

Nancy

H. P. Kramer

Attachment

CASE FILE

DO NOT PHOTOGRAPH THIS PAGE.

6/27

ANALYTICAL REFERENCE SERVICE

SAMPLE TYPE V-B (WATER, RADIOLOGICAL)

Report of a laboratory performance study conducted jointly with the participating member organizations.

Prepared by the

Analytical Reference Service

TRAINING PROGRAM

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service
Bureau of State Services
Division of Engineering Services

Robert A. Taft Sanitary Engineering Center
Cincinnati, Ohio

1961

STAFF

ANALYTICAL REFERENCE SERVICE

D. G. Ballinger, Acting Chief
R. J. Lishka, Chemist
M. C. Schneider, Chemist
D. K. Malott, Physical Science Aide
M. K. Thomas, Physical Science Aide
C. S. Sayers, Secretary

ACKNOWLEDGEMENT

We gratefully acknowledge the assistance of Theodore Jaffe, Professor of Civil Engineering, Auburn University, in the preparation and review of this report.

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FOREWORD

The Analytical Reference Service is a voluntary association of organizations responsible for the conduct of laboratory programs. It is conducted by the Training Program of the Robert A. Taft Sanitary Engineering Center for the evaluation of laboratory methods in the field of sanitary engineering. Evaluations are obtained by the cooperative efforts of the member organizations in analysis of identical samples. This cooperative mechanism provides:

1. Statistical evaluation of procedures, including precision and accuracy.

2. Opportunity for comparison of laboratory procedures and results between laboratories having similar responsibilities.

3. Exchange of information regarding weaknesses of currently approved methods.

4. Evidence of the need for further development of methods or for entirely new approaches.

5. A framework for broad-scale evaluation of new analytical methods.

Initially the program was directed toward examination of water. It has been broadened to include other types of examinations and other media.

Using the most refined grade of chemicals available, samples are designed and prepared containing measured amounts of the desired constitutents. Decisions as to qualitative makeup are made by the staff of ARS and the Center. Notice of sample availability and qualitative makeup is then sent to all member organizations.

Foreword

Immediately on receipt of the indication of willingness to participate, a portion of the sample is shipped with accompanying data forms which provide space for numerical values and a narrative critique of the analytical procedures used. The laboratories are urged to comment on the methods, including difficulties, modifications, sources of error, and other factors. Results are requested usually within sixty days.

These results, including the comments, are compiled as in the accompanying report. Examination of the data, it is hoped, will produce reliable estimates of the accuracy and precision of the methods employed.

H P Kramer

H. P. Kramer
Chief, Training Program

ANALYTICAL REFERENCE SERVICE MEMBERS*

State Agencies

Alabama Water Improvement Commission
Arizona State Department of Health
Arkansas State Board of Health
California State Department of Public Health (Berkeley)
California State Department of Public Health (Los Angeles)
California Department of Water Resources
Colorado State Department of Public Health
Connecticut State Department of Health
Florida State Board of Health (Bureau of Laboratories, Jacksonville)
Florida State Board of Health (Sanitary Engineering, Jacksonville)
Florida State Board of Health (Pensacola)
Florida State Board of Health (Winter Haven)
State of Hawaii Department of Health
Idaho Department of Health
Illinois State Department of Public Health (Chicago)
Illinois State Department of Public Health (Springfield)
Illinois State Water Survey Division
Indiana State Board of Health
Kansas State Board of Health (Sanitary Engineering Laboratories)
Kentucky State Department of Health
Louisiana State Board of Health
Maryland State Department of Health
Maryland State Water Pollution Control Commission
Massachusetts Department of Public Health
Michigan State Department of Health
Minnesota State Department of Health
Missouri Department of Public Health & Welfare
Montana State Board of Health
Nebraska State Department of Health
Nevada State Department of Health (Las Vegas)
Nevada State Department of Health (Reno)
New Hampshire Water Pollution Commission
New Jersey Department of Health
New Mexico State Department of Public Health
New York State Department of Health
New York State Conservation Department
North Carolina Department of Water Resources

*May 1, 1961

Ohio State Department of Health
Oklahoma State Department of Health
Oregon State Board of Health
Pennsylvania Department of Health
Rhode Island Department of Health
South Carolina Water Pollution Control Authority
South Dakota Department of Health
Tennessee Stream Pollution Control Board
Texas State Department of Health
Utah State Department of Health
Vermont State Department of Health
Vermont State Water Conservation Board
Virginia State Department of Health
Virginia State Water Control Board
Washington State Department of Health
West Virginia State Water Resources Commission
Wisconsin State Board of Health

Municipal Agencies

Metropolitan Water District of Southern California
Los Angeles Department of Water and Power
Los Angeles County Flood Control District
Los Angeles Department of Public Works (Hyperion Treatment Plant)
City of Pasadena
Chicago Department of Water and Sewers
Topeka, Kansas Water Works
Flint Water Plant, Department of Public Works and Utilities
St. Louis County Water Company
St. Louis Department of Health and Hospitals
Metropolitan Utilities District (Omaha, Nebraska)
City of Durham Department of Water Resources (North Carolina)
Dayton Department of Service and Buildings
Philadelphia Suburan Water Company
Philadelphia Water Quality Control and Research Division

Federal Agencies

Food and Drug Administration, Division of Pharmacology, Washington, D.C.
Tennessee Valley Authority, Wilson Dam, Alabama (Occupational Health Branch)
Tennessee Valley Authority, Wilson Dam, Alabama (Air Pollution Control Section)
Tennessee Valley Authority, Chattanooga, Tennessee (Stream Pollution Control)

2794th U.S. Air Force Dispensary - Class B
U.S. Army Environmental Hygiene Agency, Maryland
Sanitary Engineering Division, 13th Naval District, Seattle, Washington
U.S. Dept. of the Interior, Geological Survey, Sacramento, California
U.S. Dept. of the Interior, Geological Survey, Denver, Colorado
U.S. Dept. of the Interior, Geological Survey, Washington, D.C.
U.S. Dept. of the Interior, Bureau of Reclamation, Denver, Colorado
U.S. Dept. of the Interior, Geological Survey, Columbus, Ohio
U.S. Dept. of the Interior, Geological Survey, Philadelphia, Pennsylvania

Universities

Case Institute of Technology, Dept. of Civil and Sanitary Engineering
Columbia University
Georgia Institute of Technology, Dept. of Applied Biology
University of Pittsburgh, Graduate School of Public Health
Purdue University, Department of Chemistry
Rutgers University, Department of Sanitation

Industry

American Cyanamid Company, Bound Brook, New Jersey
Battelle Memorial Institute, Columbus, Ohio
California Water Service Company, San Jose, California
Central Farmers Fertilizer Company, Montpelier, Idaho
Dearborn Chemical Company, Chicago, Illinois
E.I. du Pont de Nemours & Company, Aiken, South Carolina
Ekroth Laboratories, Inc., Brooklyn, New York
Food Machinery & Chemical Corp., Pocatello, Idaho
Goodyear Atomic Corporation, Portsmouth, Ohio
Industrial Chemicals, Inc., South Bend, Indiana
Monsanto Chemical Company, St. Louis, Missouri
NALCO Chemical Company, Chicago, Illinois
Radiation Detection Company, Palo Alto, California
Reynolds Electrical & Engineering Co., Las Vegas, Nevada
Roy F. Weston, Inc., Newtown Square, Pennsylvania
Sandia Corporation, Sandia Base, Albuquerque, New Mexico
Tracerlab, Inc., Richmond, California

Foreign

Vancouver, B.C., Department of National Health & Welfare
Ottawa, Ontario, Department of National Health & Welfare

PARTICIPANTS IN THIS STUDY

Alabama Water Improvement Commission
Arkansas State Board of Health
California State Department of Public Health (Berkeley)
California Department of Water Resources
Connecticut State Department of Health
Florida State Board of Health (Sanitary Engineering, Jacksonville)
State of Hawaii Department of Health
Illinois State Department of Public Health (Chicago)
Illinois State Department of Public Health (Springfield)
Illinois State Water Survey Division
Kentucky State Department of Health
Michigan State Department of Health
Minnesota State Department of Health
Montana State Board of Health
New Hampshire Water Pollution Commission
New Jersey Department of Health
New York State Department of Health
North Carolina Department of Water Resources
Ohio State Department of Health
Pennsylvania Department of Health
Rhode Island Department of Health
South Carolina Water Pollution Control Authority
Tennessee Stream Pollution Control Board
Utah State Department of Health
Vermont State Water Conservation Board
Virginia State Department of Health
Virginia State Water Control Board
Wisconsin State Board of Health
Metropolitan Water District of Southern California
Los Angeles Department of Water and Power
Chicago Department of Water and Sewers
Philadelphia Water Quality Control and Research Division
U.S. Army Environmental Hygiene Agency, Maryland
U.S. Dept. of the Interior, Geological Survey, Denver, Colorado
U.S. Dept. of the Interior, Geological Survey, Columbus, Ohio
Case Institute of Technology, Dept. of Civil and Sanitary Engineering
University of Pittsburgh, Graduate School of Public Health
Rutgers University, Department of Sanitation
Sandia Corporation, Sandia Base, Albuquerque, New Mexico
Southeastern Radiological Health Facility, PHS, DHEW
Southwestern Radiological Health Facility, PHS, DHEW

SAMPLES ON WHICH REPORTS HAVE BEEN COMPLETED

SAMPLE TYPE I AND I-A

(Water, Minerals) analyzed for calcium, magnesium, hardness, sulfate, chloride, alkalinity, nitrite, nitrate, sodium and potassium.

SAMPLE TYPE II

(Water, Heavy Metals) analyzed for copper, cadmium, aluminum, chromium, iron, manganese, and zinc.

SAMPLE TYPE III

(Water, Fluoride) analyzed for fluoride in the presence and absence of interferences with and without distillation, using a specified procedure.

SAMPLE TYPE IV

(Air, Inorganics) analyzed for chloride, sulfate, fluoride and nitrate in aqueous solution and on glass fiber hi-vol filter mats.

SAMPLE TYPE V

(Water, Radiological) analyzed for gross beta activity.

SAMPLE TYPE VI

(Water, Surfactant) analyzed for surfactant in various waters.

SAMPLE TYPE VII

(Water, Oxygen Demand) analyzed for biochemical oxygen demand and chemical oxygen demand.

SAMPLES NOW UNDER STUDY

(Water, Fluoride) analyzed for fluoride in the presence and absence of interferences with and without distillation, using a specified procedure.

(Water, Minerals, Number 3) analyzed for calcium, magnesium, hardness, sulfate, chloride, alkalinity, nitrite, nitrate, sodium and potassium.

(Air, Lead, Number 1) analyzed for lead.

SAMPLE TYPE V-B (WATER, RADIOLOGICAL)

PURPOSE OF STUDY SERIES

Governmental agencies are increasingly concerned with the determination of radioactivity concentrations in the environment (air, food, milk, water, vegetation). Samples routinely are measured for gross activity and, where indicated, for the activity of specific radionuclides. Measurement of gross activity is complicated by many factors, such as the calibration standard used, sample preparation, self-absorption, and others. To evaluate certain of these complicating factors the Analytical Reference Service undertook a series of studies on the determination of radioactivity levels of environmental samples.

STUDY BACKGROUND

The Analytical Reference Service sent out the sample of this series to 31 participating groups in August 1958. The results of the study were published in the report, "Sample Type V, Water, Radiological", June, 1959.

Using Thallium^{204} (a pure beta-emitting radionuclide) in tap water, this study indicated that the determination of gross beta activity was affected by the following factors:

1. solids' drying temperature
2. type of counter
3. choice and preparation of standard
4. sample volume

OBJECTIVES OF THIS STUDY

In view of the results, it was decided to investigate further the effects of these four factors when the environmental sample contained a beta- and gamma-emitting radionuclide. Specific objectives of the second study were to evaluate:

1. Control of variables by a recommended sample-preparation procedure
2. Type of counter for beta and/or gamma counting
3. Control of standard preparation
4. Effect of sample thickness (self-absorption)

ANALYSIS SCHEME

All laboratories participating in this study were asked to process the reference sample in their usual manner with the following recommended stipulations:

1. Volume of sample — 500 ml
2. Evaporation temperature — 95° - 98°
3. Evaporation-container rinse
   - a. acid — HCl for aluminum planchets; HNO_3 for stainless steel planchets
   - b. concentration — 0.1N
   - c. volume — < 0.5 ml (several drops)
4. Final drying — 105°C for 2 hours (oven)
5. Analysis in triplicate was recommended

SAMPLE PREPARATION

In the report on Sample Type V, the use of Thallium^{204} as a standard for gross beta activity measurements of mixtures of unknown radionuclides in water samples was questioned. Cesium^{137} was suggested as a replacement, one advantage being its long half-life (30 years). Because of this characteristic, among others, it was decided to use this radionuclide in the second reference sample in the series. However, this study is not intended to evaluate Cs^{137} against Tl^{204} as a counting standard.

To simulate a natural water, this sample, containing Cesium^{137}, was made with tap water. Aliquots of the sample were distributed to 45 participating agencies together with the above suggested procedure. Accompanying each sample was a report form requesting information concerning methods of sample preparation, counting techniques, and other factors. The data obtained from these forms comprise the basis for this report.

The sample was prepared by adding ten lambdas (microliters), accurately measured, of a Cs^{137} solution to 320 liters of Cincinnati tap water. The Cs^{137} solution was calibrated by the supplier at 6.29×10^5 gamma emissions per second per milliliter. No decay correction was considered necessary because of the 30-year half-life of Cs^{137}. Uniformity of the Cs^{137} distribution throughout the reference solution was determined by a procedure developed by the Physical and Instrumental Analyses Group of the Radiological Health Research Activities of SEC.

Three 3.5-liter aliquots of the Type V-B sample were taken, one each from the top of the mixing tank, from the middle, and from the bottom. Each was counted for 100 minutes on a low background, 100-channel analyzer using an inverted well-type beaker over a four-inch, NaI detection crystal. The results of the gamma scans showed that the activities of the three aliquots, in terms of counts per minute under the photopeak, agreed within 2%. These data were then calculated as the gamma activity, expressed as $\mu c/ml$ ($\times 10^{-7}$). The apparent beta activity, in $\mu c/ml$ ($\times 10^{-7}$), was obtained by multiplying the gamma activity by 1.33.

Use of the term "apparent beta activity" may require clarification. When using Cs^{137} as a standard, it has been observed that the beta activity of standard solutions was higher than expected, calculated on the basis of the calibrated gamma activity. This anomaly is due to the fact that Ba^{137m} (the radioactive daughter of Cs^{137}) emits conversion electrons as well as gamma photons. The conversion electrons are counted as beta particles while the gamma photons are not detected. As a result, the beta emission rate is higher than expected, while the gamma emission rate is lower. For example, for every 100 disintegrations of Cs^{137}, 109.7 beta counts and 82.3 gamma counts could be detected, assuming 100% counting efficiency. This counting of the conversion electrons together with the beta particles leads to an "apparent beta activity".

Assuming that the gamma emission rates are known, the following correction has to be made to calculate the apparent beta activity (beta particles plus conversion electrons):

$$\text{Apparent } \beta \text{ activity} = \frac{109.7}{82.3} \times \text{gamma emission rate}$$

$$= 1.33 \times \text{gamma emission rate}$$

This procedure served as a check on the distribution uniformity of the Cs^{137}, as well as a gamma calibration of the Type V-B sample. Another calibration check was obtained on a sample sent to Oak Ridge National Laboratory for beta analysis. Results of these analyses are given in Table I.

Table I. STANDARDIZATION OF SAMPLE TYPE V-B

	Beta (μc/ml)	Gamma (μc/ml)
Calculated activity	7.1×10^{-7}	5.3×10^{-7}
Rad. Health Research Activities	6.8×10^{-7}	5.1×10^{-7}
O. R. N. L.	6.3×10^{-7}	---
Average (activity added)	6.7×10^{-7}	5.2×10^{-7}

REPORTED DATA

Forty-one of the 45 groups participating in the study returned results in time to be included in this report. Agencies #24 and #27 submitted two results each and both were assigned double codes - 24A and 24B and 27A and 27B respectively. All of the data from the 41 reporting agencies are summarized in Table II and graphed in Figure 1.

Table III summarizes the beta counting as to (1) type of counter used, (2) choide of standard, (3) sample thickness. The data on which this summary is based are arrayed in Table IV. Table V presents the factors affecting the gamma results.

TABLE II. SUMMARY OF DATA

BETA COUNTING

Number of agencies reporting	41
Number of results	43
Amount of radioactivity in sample, mean value	6.7×10^{-7} µc/ml
Amount recovered, mean value	5.8×10^{-7} µc/ml
Median	6.4×10^{-7} µc/ml
Range	$0.2 - 8.2 \times 10^{-7}$ µc/ml
50% range	$\pm 0.8 \times 10^{-7}$ µc/ml
Number of results within ± 10% of amount added	25
Number of results within ± 50% of amount added	28
Mean % deviation from amount added	18
Mean % deviation within laboratories	4
Standard deviation from mean	1.6
Standard deviation from amount added	1.8

(Agencies #15 and #32 not included. Reported only one result).

GAMMA COUNTING

Number of results	8
Amount of radioactivity in sample, mean value	5.2×10^{-7} µc/ml
Mean of all results	5.8×10^{-7} µc/ml
Range	$1.4 - 11.0 \times 10^{-7}$ µc/ml
Number of results within ± 10% of amount added	1
Number of results within ± 50% of amount added	5

Table III. SUMMARY OF FACTORS AFFECTING BETA-COUNTING RESULTS

	Radioactivity (Mean $\bar{x}$) $\times 10^{-7}$ μc/ml
TYPE OF COUNTING INSTRUMENT	
End Window (9 results)	5.8
1) Gas flow end window (6 results)	5.7
2) G.M. end window (3 results)	6.1
Internal proportional (34 results)	5.8
CHOICE OF STANDARD	
Thallium^{204} (17 results)	6.2
Cesium^{137} (5 results)	5.8
Carbon^{14} (7 results)	6.0
Other (14 results)	5.2
SAMPLE THICKNESS	
Less than 20 MG/CM^2 (31 results)	6.3
Greater than 20 MG/CM^2 (7 results)	4.5
Not given (5 results)	5.2

Table IV. ARRAY OF FACTORS AFFECTING RESULTS

Agency No.	Amount reported as beta $\times 10^{-7}$ μc/ml	Beta calibration standard	Type of Instrument	Average solids thickness in mg/cm^2
29	0.2	Co^{60}	I.P.C.	35
38	2.2	Bi^{210}	I.P.C.	40
18	3.1	Bi^{210}	Gas Flow E.W.	24
26	3.1	Ra D&E	I.P.C.	--
35	3.4	C^{14}	I.P.C.	11.6
39	4.5	Tl^{204}	I.P.C.	26.4
36	4.6	Tl^{204}	I.P.C.	10.3
32	4.8	Cs^{137}	Gas Flow E.W.	--
40	4.8	Cs^{137}	I.P.C.	9.6
2	5.1	Uranium Oxide	I.P.C.	5.4
21	5.2	Tl^{204}	G.M.	14.0
9	5.3	C^{14}	I.P.C.	9.6
24B	5.3	Y^{90}	Gas Flow E.W.	
25	5.4	Tl^{204}	I.P.C.	9.5
7	5.5	Tl^{204}	I.P.C.	10.2
10	5.6	Tl^{204}	I.P.C.	11.1
11	5.9	C^{14}	I.P.C.	9.5
4	6.1	C^{14}	I.P.C.	10.1
24A	6.2	Y^{90}	Gas Flow E.W.	--
41	6.3	Cs^{137}	I.P.C.	17.7
33	6.3	Cs^{137}	G.M.E.W.	42.1
17	6.4	Tl^{204}	I.P.C.	5.1

Table IV. ARRAY OF FACTORS AFFECTING RESULTS (Cont'd)

Agency No.	Amount reported as beta $\times 10^{-7}$ μc/ml	Beta calibration standard	Type of Instrument	Average solids thickness in mg/cm^2
23	6.4	Tl^{204}	I.P.C.	10.2
8	6.4	Tl^{204}	I.P.C.	23
22	6.5	RHHB ABS Curve	I.P.C.	8.5
15	6.5	Sr^{90}	I.P.C.	---
6	6.6	Tl^{204}	I.P.C.	9.6
13	6.6	C^{14}	I.P.C.	12.9
5	6.6	Uranium Oxide	I.P.C.	12.6
16	6.6	Tl^{204}	I.P.C.	12.6
12	6.6	Tl^{204}	I.P.C.	8.4
30	6.7	Cs^{137}	G.M.E.W.	19
3	6.9	Tl^{204}	I.P.C.	11.3
1	6.9	---	I.P.C.	9.4
37	6.9	Tl^{204}	I.P.C.	16.2
19	7.0	Tl^{204}	I.P.C.	11.0
34	7.2	---	I.P.C.	12.0
20	7.2	C^{14}	I.P.C.	11.0
27B	7.2	Tl^{204}	Gas Flow E.W.	10.5
28	7.3	Ra D&E	I.P.C.	10.4
31	7.4	Ra D&E	I.P.C.	11.3
14	7.4	C^{14}	I.P.C.	13.1
27A	7.7	Tl^{204}	Gas Flow E.W.	20.7

Table V. ARRAY OF GAMMA RESULTS

Agency No.	Amount reported as gamma $\times 10^{-7}$ μc/ml	Gamma calibration standard	Type of instrument
16	1.4	Co^{60}	Scintillation
34	2.6	Cs^{137}	Single Channel Scintillation Spectrometer
32	4.2	Cs^{137}	Scintillation
33	5.3	Cs^{137}	Scintillation Spectrometer Bulk Analyzer
41	6.2	Cs^{137}	Scintillation
36	6.3	Cs^{137}	G. M. End Window 1 GM/CM^2 Aluminum sample cover
28	9.2	Cs^{137}	Scintillation Spectrometer
30	11.0	Cs^{137}	G. M. End Window

DISCUSSION OF BETA COUNTING DATA

In general, the counting of the Type V-B sample for beta emissions seems to have been affected by:

1. type of counting instrument
2. choice of standard
3. sample thickness

Type of Counting Instrument

From the results, it appears that end-window counters can be as reliable as internal proportional counters, although the previous study (Type V) indicated lower counting values for the end-window types. Since the importance of positioning the standard and sample with the end-window counter was pointed out in the report of the first study, the improvement noted in this study may be due, at least in part, to more careful positioning. It should be re-emphasized that reproduction of geometry is of prime importance in end-window counting.

While positioning is not so important with the internal proportional counter, it is important that samples be completely dry. Vapors, such as moisture or acid, if mixed with the counting gas, produce erratic counting.

Choice of Standard

The choice of a calibration standard appears to be an influential factor in the counting of environmental samples. Fourteen of the 41 laboratories used a standard other than Tl^{204}, Cs^{137}, or C^{14}. Although no calibration standard was suggested in this study procedure, the mean value of the results of the fourteen laboratories was lower than the mean value for any of the other three standards used. However, insufficient data precludes incriminating any of these materials.

As expected, a number of laboratories used Tl^{204} as a beta standard for calibrating their counting instruments. This radionuclide has served as a gross beta standard for interpreting radiological counting data from water samples for several years, but its use in this study created an unusual problem. The half-life of Tl^{204} has long been uncertain. At the time the standards were purchased by the majority of the participating agencies, the half-life was given as 4.1, 4.0, 3.5, 2.7, or 2.5 years, depending on the supplier. The use of this range of values in the calculation of beta activity at the time of instrument standardization resulted in errors in efficiency factors. These errors were carried over to the beta activity values reported on the Reference Samples. For the past five years the Oak Ridge National Laboratory has been following the decay of Tl^{204} standards commercially available. The half-life of these standards has been calculated at 3.56 years. Since the half-life values used by the participating agencies were significantly different, it was necessary to correct the reported beta activity before statistically analyzing the data.

Correction of the beta activities was calculated by use of the formula:

$$\frac{\text{Activity reported}}{\text{Corrected activity}} = \frac{e^{\frac{-0.693}{T\frac{1}{2}\text{ used}}t}}{e^{\frac{-0.693}{3.56}t}}$$

where t = elapsed time between initial standardization and sample analysis.

The corrected values are shown in Table VI.

Table VI. THALLIUM-204 HALF-LIFE CORRECTIONS

Agency No.	$T_{\frac{1}{2}}$ used	Beta activity reported ($\times 10^{-7}$ μc/ml)	Corrected beta activity ($\times 10^{-7}$ μc/ml)
17	4.1	7.4	6.4
27B	4.1	7.7	7.2
27A	4.1	8.2	7.7
8	4.1	6.8	6.4
6	4.1	7.0	6.6
39	4.1	4.7	4.5
12	4.0	7.0	6.6
36	4.0	4.9	4.6
16	4.0	7.0	6.6
37	4.0	7.1	6.9
21	4.0	5.5	5.2
25	3.5	5.3	5.4
10	3.5	5.5	5.6
23	3.5	6.4	6.5
7	2.7	4.7	5.5
3	2.7	5.9	6.9
34	2.5	5.9	7.2

Five of the agencies chose Cs^{137} as a standard and obtained a mean result of 5.8×10^{-7} μc/ml for the reference sample. It would be expected that the best results for Sample V-B would be obtained from the use of efficiency factors determined from a self-absorption curve representing various thickness of solids with equal amounts of Cs^{137} activity added. Since so few labs used Cs^{137} as a standard, this information was not forthcoming.

Two of the laboratories, 33 and 41, did employ such a procedure and each obtained results of 6.3×10^{-7} μc/ml. Agency 41 processed the Type V-B sample and then prepared Cs^{137} self-absorption standards by mixing known amounts of Cs^{137} with an amount of K_2SO_4 equal to the weight of the solids on the sample planchets. Agency 33 prepared similar standards using CaO. Although a G.M. end-window counter and a thick sample were used, this laboratory obtained a reliable counting value.

The seven agencies Using C^{14} as a standard obtained results having a mean value of 6.0×10^{-7} μc/ml. However, it should be emphasized that the low beta energy of C^{14} (0.155 Mev) renders this standard unsuitable

for self absorption studies because of the large absorption of this low beta energy in any appreciable amount of solid materials. Also, the beta energy of C^{14} does not represent the average beta energy of mixed fission products commonly found in environmental samples.

Effect of Sample Thickness

In general, the best results were obtained by agencies using a sample thickness of less than 20 mg/cm^2. The mean activity for 31 agencies reporting a thickness less than 20 mg/cm^2 was 6.3×10^{-7} μc/ml. Of further interest is the fact that only two of the 25 agencies comprising the 50% range (from 6.1 to 7.4×10^{-7} μc/ml) reported sample thickness values greater than 20 mg/cm^2. The mean for 7 results greater than 20 mg/cm^2 thickness was 4.5×10^{-7} μc/ml. For 5 agencies which did not report a sample thickness, the mean was 5.2×10^{-7} μc/ml.

One possible reason for the "high" sample thickness obtained by some of the laboratories was the 500-ml sample volume requested by the Analytical Reference Service. A better method might have been to suggest the use of 500 ml by agencies using 2" diameter planchets and 250 ml or less by agencies using 1" diameter planchets.

DISCUSSION OF GAMMA COUNTING DATA

The gamma results are presented in Table V. Since there were only 8 gamma results, no attempt was made to apply the regular statistical calculations.

The main factors which seemed to affect the gamma determinations were:

1. use of a G.M. end-window counter
2. use of a single channel analyzer to determine a specific radionuclide, instead of performing a gross gamma count.

The efficiency of a G.M. end-window counter for gamma detection is very low, introducing a very large error into the calculation of radioactivity concentrations.

Some confusion seemed to exist among the participating agencies as to whether the V-B sample should be analyzed for gross activity or for a specific radionuclide, Cesium 137.

Although the Type V-B sample had been standardized in terms of Cs^{137} activity, it would have been more desirable for the laboratories to report their answers in terms of gross activity or gross activity and Cs^{137} activity, if desired. This procedure, which may have produced results on the "high" side due to an apparent higher counting efficiency, still would be more desirable from the public health standpoint. Therefore, the use of a single channel analyzer for "gross" counting may be quite satisfactory.

The use of a single channel analyzer for gamma analysis of specific radionuclides in environmental samples has definite limitations. Due to the relatively low amount of radioactivity contained in an environment sample (such as Type V-B) an excessive amount of time is consumed in checking each channel to obtain enough activity to achieve good counting statistics. The use of a multi channel analyzer greatly reduces counting time and counting errors for specific analysis of gamma emitting radionuclides.

SUMMARY OF RESULTS

From the data submitted by 41 participating agencies for gross beta and/or gamma radioactivity determinations on an environmental sample (Water, Radiological, Type V-B) containing Cesium 137 as the radionuclide, the following conclusion was reached:

1. With care in positioning the standard and the sample in respect to the end-window counter, equally reliable results for gross beta counting were obtained by using either the end-window or the internal proportional type.
2. For gamma counting, the use of the G. M. end-window counter may result in large errors in radioactivity concentration due to its very low efficiency. If gross gamma counting is desired, the single channel analyzer appears to be satisfactory. For the analysis of specific gamma emitting radionuclides, the use of a multi channel analyzer reduces counting time and counting errors.

3. It appears that Thallium 204 is satisfactory as a standard for instrument calibration for gross beta counting of water samples. The use of self-absorption correction curves for samples containing solids should produce more reliable results. Although good values may be obtained with C^{14} as a standard, it has two disadvantages:

   (a) large absorption in the presence of solids, and

   (b) energy not representative of the average beta energy of mixed fission products.

4. Sample volumes chosen to give a counting sample thickness of 20 mg/cm^2, or less, gave the best results for gross beta counting.

COMPARISON OF SAMPLE TYPE V AND V-B

Table VII compares the results obtained in the current study, Type V-B, and the previous study, Type V, in the Water, Radiological series. On the basis of this data analysis, there appears to be an over-all improvement in the results of the gross beta counting, as shown by the greater numbers of results within any given percentage of the amount of radioactivity added. Further, the mean recovery of the added radioactivity in the V-B sample was 86.6%, whereas for the V sample it was 76.0%.

From this table it can be seen that both sets of data have the same deviation around their respective mean values. This similarity indicates that more attention should be given to the factors involved in gross beta counting of water samples.

One disturbing fact, from a public health viewpoint, brought out by these studies, is the number of agencies reporting concentrations less than that percent in the sample. In the Type V study, 80% of the agencies reported activity concentrations equal to or less than the added amount; in the V-B study, 63% fell into this group. These percentages represent an improvement but still leave much to be desired.

Twenty-one of the agencies participating in the first study of this series took part in this second study after receiving the report and data analysis for the first study. The major differences between these two studies were:

1. The radionuclide in the first sample was a pure beta emitter, Tl^{204}; the second was both a beta and gamma emitter, Cs^{137}.

2. A sample preparation procedure was suggested, with drying temperature, sample volume, etc. stated.

3. The laboratories were aware of factors influencing the determination of gross beta activity since these were noted in the first report.

Table VIII summarizes the results from the 21 laboratories for each of the two studies. It is apparent that these results show an improvement in the determination of the gross beta activity in the second sample. It might be assumed that these better results are due to more careful techniques, since the first report (Type V) pointed out some of the factors influencing beta counting determinations. If this is so, it may be that the similarity for all Type V-B and Type V results is due, at least partially to inattention to techniques by other labs in the V-B study who participated for the first time.

From the data in Table VIII, several observations are warranted:

1. The mean beta value of the V-B sample for these 21 agencies was 6.2×10^{-7} μc/ml or 92.5% of the radioactivity added. For the entire group (41 agencies) the mean value was 5.8×10^{-7} μc/ml or 86.6% of the radioactivity added. The 21 agencies on the first sample achieved only 76.0% recovery.

2. The results of these 21 labs in the V-B study have a smaller standard deviation from the mean, indicating an overall improvement in gross beta counting.

3. Of these 21 agencies, 12 labs (57%) reported results equal to, or less than, the activity added to the reference sample. While these results are better than for the group as a whole, and much better than in the Type V study, further improvement is desirable.

Table VII. COMPARISON OF DATA ON RADIOLOGICAL SAMPLES V AND V-B FROM ALL AGENCIES

	Samples	
	V	V-B
Number of agencies reporting	25	41
Number of results	26	43
Amount of radioactivity in sample $\mu c/ml \times 10^{-7}$		
As beta	5.44	6.7
As gamma	----	5.2
Amount of beta radioactivity recovered, mean $\mu c/ml \times 10^{-7}$	4.13	5.8
Percent of beta radioactivity recovered	76.0	86.6
Number of results within $\pm$ 10% of amount added	8	22
Percent of total no. of results	31	51
Number of results within $\pm$ 50% of amount added	20	39
Percent of total no. of results	77	91
Standard deviation from mean	1.8	1.6
Standard deviation from amount added	2.3	1.8
50% range	$\pm$0.8	$\pm$0.7

Table VIII. DATA SUMMARY FROM AGENCIES PARTICIPATING IN BOTH TYPE V AND V-B RESULTS

	Type V	Type V-B
Number of results	21	21
Amount of beta activity in sample, μc/ml	5.44×10^{-7}	6.7×10^{-7}
Amount recovered, mean, μc/ml	4.1×10^{-7}	6.2×10^{-7}
Amount recovered, percent	75.7	92.5
Standard deviation from mean recovery	1.93	1.23

GENERAL SUMMARY AND CONCLUSIONS

As a result of a second sample, Type V-B, in the Water, Radiological study series of the Analytical Reference Service, certain conclusions can be stated regarding gross beta and/or gamma radioactivity determinations on an environmental water sample:

1. Sample preparation drying temperatures must be chosen to preculde any loss of standard or sample due to volatilization. If Tl^{204} is used as a standard, temperatures must be kept below 125°C.

2. Volumes should be chosen to keep the counting sample thickness to a minimum, preferably below 20 mg/cm^2, to reduce self-absorption losses.

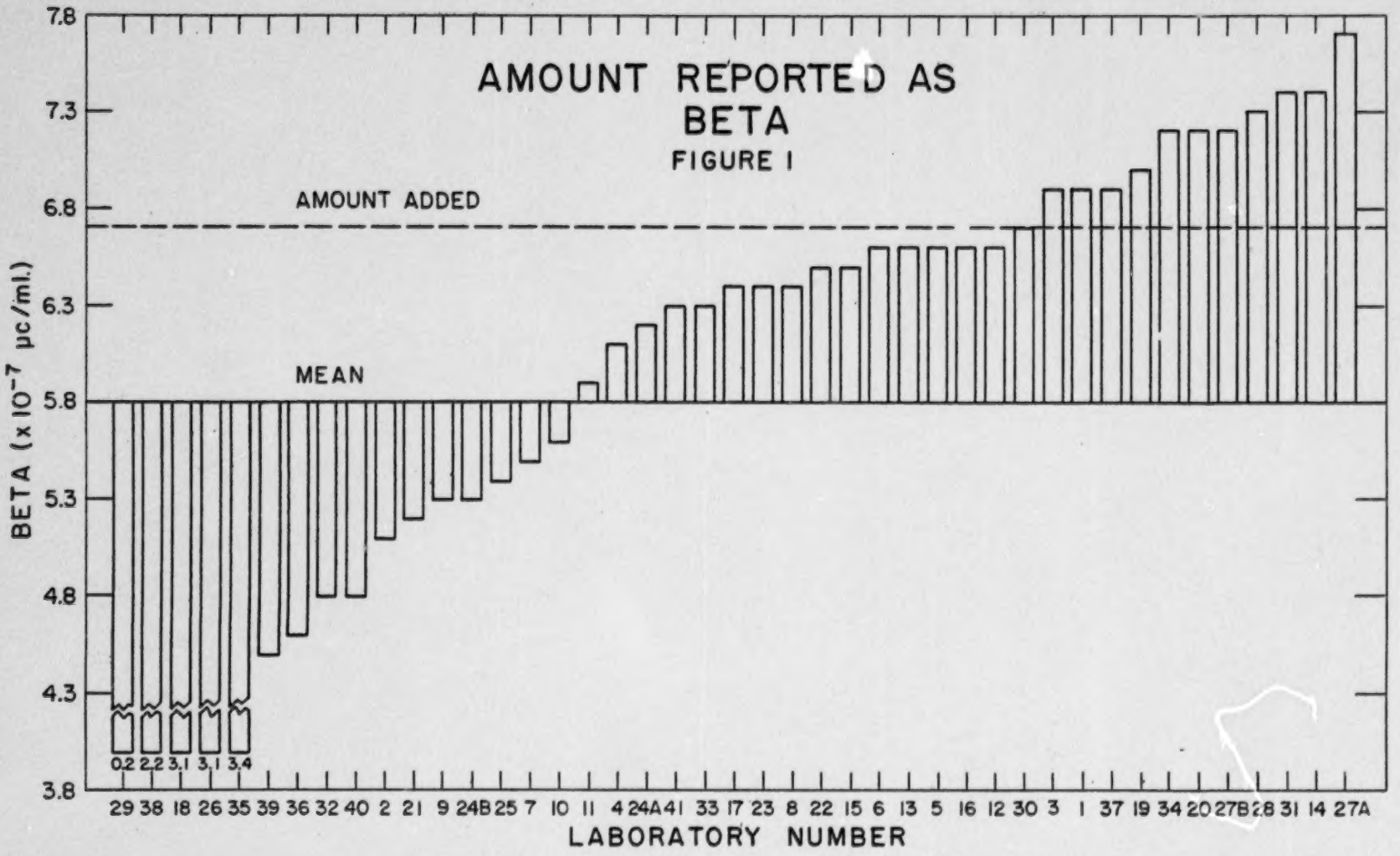
3. With careful attention to positioning (geometry) of standard and sample, end-window counters are as satisfactory as internal proportional counters for gross beta counting.

4. For gamma counting, end-window counters do not give as reliable results as channel analyzers. For gross gamma determinations, single channel instruments are satisfactory; for specific gamma emitting nuclide determinations multi channel analyzers are preferable.

5. Tl^{204} may be used as a calibration standard for gross beta counting, provided the above precautions are observed. In spite of the theoretical advantages of Cs^{137}, sufficient data are not available at this time to evaluate Cs^{137} against Tl^{204}.

6. When the results submitted by the agencies who participated in both studies are compared, much improvement in beta counting is noted. It is likely that this improvement can be attributed to careful attention, in the second study, to the influential factors pointed out in the first study.

7. Radioactivity determinations in environmental water samples appear to yield low results. The significance of this in public health fields is apparent.



ANALYTICAL REFERENCE SERVICE
SAMPLE TYPE V-B
(WATER, RADIOLOGICAL)